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Using activation method to measure neutron spectrum in an irradiation chamber of a research reactor*

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Neutron spectrum should be measured before test samples are irradiated. Neutron spectrum in an irradiation chamber of a research reactor was measured by using activation method when the reactor is in normal operation under 2 MW. Sixteen kinds of non-fission foils (19 reaction channels) were selected, of which 10 were sensitive to thermal and intermediate energy regions, while the others were of different threshold energy and sensitive to fast energy regions. By measuring the foil radioactivity, the neutron spectrum was unfolded with the iterative methods SAND-II and MSIT. Finally, shielding corrections of group cross-section and main factors affecting the calculation accuracy were studied and the uncertainty of solution was analyzed using the Monte Carlo method in the process of SAND-II.

Keywords: Neutron spectrum, Iterative method, SAND-II, MSIT, Group cross-section

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I. INTRODUCTION

Most neutrons in Irradiation Chamber 3 of the research reactor are intermediate and fast neutrons, because most thermal neutrons are stopped in the lead box. Many samples can be irradiated in the irradiation chamber. Measuring neutron spectrum at the irradiation point can provide important information for irradiation experiments [1].

Activation method is a simple and widely used method for measuring neutron energy spectrum [2–4]. The foil size is small enough to ensure a uniform neutron field at the measuring point. Different foil materials can measure neutrons in different energies. Activation method depends greatly on the spectrum unfolding technology. In recent years, many unfolding methods have been developed. They can be divided into three classes: [5, 6] iteration method, method of undetermined coefficients, Monte Carlo method. The method of undetermined coefficients was once widely used, but it produces rough results. Monte Carlo method dealing with a large number of neutron energy groups shall request great deal of computation time, while reliability of the results is poor when fewer number of activation foils was chosen. In recent years, with the development of computer technology and the improvement of the cross section library, iteration method has been widely used, with much more accurate results. SAND-II and MSIT are both iteration methods. In this work, MC uncertainty analysis was added to SAND-II, so as to obtain even better results.

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II. THEOETICAL ANALYSIS

A. The fundamental procedures of activation method

The fundamental of activation method for measuring neutron spectrum is as follows: a group of foils, with known activation of sections of the nuclides, are irradiated at the measuring point, and the relationship between single reaction rate and neutron energy spectrum is given by Eq. (1) [7]:

$$A_i = \int_0^\infty \phi(E)\sigma_i(E)\mathrm{d}E \quad (i = 1, 2 \ldots n), \tag{1}$$

where A_i is single nuclear reaction rates of foil i, $\phi(E)$ is neutron energy spectrum, $\sigma_i(E)$ is nuclear reaction cross section of foil i at neutron energy E, n is the number of adopted nuclear reaction channels. Stable nuclei are activated by neutron irradiation, and A_i is obtained by measuring γ-rays coming from activated nucleus. $\sigma_i(E)$ can be obtained from an evaluation of the database. $\phi(E)$ can be obtained by solving the Eq. (1) in theory. The process of finding the solution $\phi(E)$ from Eq. (1) is called as unfolding spectrum.

B. Basic principle of spectrum unfolding

Due to limited number of activation foils in the measurement, one can use the A_i data and obtain an approximate $\phi(E)$, rather than a continuous spectrum of $\phi(E)$, with some assumptions for neutron field and restriction conditions.

In the iterative methods of SAND-II and MSIT, the neutron spectrum unfolding is done by dividing the energy region into many energy groups, Eq. (1) could be transformed into Eq. (2):

$$A_i = \sum_{j=1}^{m} \phi_j(E)\sigma_{i,j}(E)\Delta E_j \qquad (i = 1, 2 \ldots n) \qquad (2)$$

where $\phi_j(E)$ is the average neutron flux density of energy group i; $\sigma_{i,j(E)}$ is the average nuclear reaction cross section of energy group j for activation foil i; ΔE_j is the energy interval of energy group j; m stands for the number of energy group.

The initial neutron energy spectrum $\phi_j^{[0]}(E)$ is obtained based on the conditions of the neutron field. The single nuclear reaction rate A_i^0 could be calculated if $\phi_j^{[0]}(E)$ and group cross sections are known. The corrected spectrum of iteration [K+1] can be obtained by using correction factor and the spectrum of iteration[K] for both SAND-IIand MSIT. Eqs. (3) and Eq. (4) [7] are modifier formulas for SAND-II and MSIT, respectively.

$$\phi_j^{[K+1]}(E) = \phi_j^{[K]}(E)exp(C_j^{[K]}), \qquad (3)$$

where, $\phi_j^{[K]}$ and $\phi_j^{[K+1]}$ are energy spectra of iteration [K] and [K+1], respectively; $C_j^{[K]} = \sum_{i=1}^{N} W_{i,j}^{[K]} \ln R_i^{[K]} / \sum_{i=1}^{N} W_{i,j}^{[K]}$ is the correction factor, $W_{i,j}^{[K]} = A_{i,j}^{[K]}/A_i^{[K]}$ is the weighting factor, $R_i^{[K]} = A_i/A_i^{[K]}$ is the ratio of measured value and calculated value.

$$\phi_j^{[K+1]}(E) = \phi_j^{[K]}(E)C_j^{[K]} \qquad (4)$$

where $C_j^{[K]} = \sum_{i=1}^{N}(\sigma_{i,j}/A_i^{[K]}) / \sum_{i=1}^{N}(\sigma_{i,j}/A_i)$ is the correction factor of energy interval j.

The iterative process stops when one of the three termination conditions are met: (1) the standard deviation between the calculated reaction rate and the measured reaction rate is less than a given value; (2) all the difference between the two adjacent iterative differential spectrum for each energy group is less than a given value; (3) the number of iterations reaches a given value.

III. EXPERIMENTAL MEASUREMENT OF REACTION RATE

Sixteen kinds of activation foils were adopted in the experiment. The diameter of Mg and Mo foils is 8 mm, while the other foils are of 20 mm diameter. The characteristic parameters of activation foils and measurement data of the single nuclear reaction rate are shown in Table 1. After proper cooling, γ-ray spectra of the irradiated foils were measured by high-purity Ge detectors with the detection efficiency scaled in advance. Eq. (5) is used to calculate the single reaction rate [8].

$$A_i = \frac{C\lambda_i}{\gamma_d \varepsilon N_i (1 - e^{-\lambda_i t_0}) e^{-\lambda_i (t_1 - t_0)} (1 - e^{-\lambda_i (t_2 - t_1)})} \qquad (5)$$

where, C is net peak area of the γ-ray spectrum; γ_d is the branching ratio of γ-ray; ε is the detection efficiency; m_i is the mass of foil i; $N_i = (m_i/M_i)\theta_i \times 6.023 \times 10^{23}$ is the number of nucleus for foil i, M_i is the isotopic atomic weight of nucleus i, θ_i is the content of isotope for nucleus i; λ_i is decay constant of nucleus i; t_0 is irradiation time; $t_1 - t_0$ is cooling time; $t_2 - t_1$ is measuring time.

IV. RESULTS

A. Process of group cross section

The cross-section data for the spectrum unfolding are from ENDF/B-VII. They are treated in the following processes. First, the complete raw data is selected from ENDF using Prepro and transformed to linearization. The next procedure is to remake the resonances. Then, Doppler broadening was done based on desired temperature. Finally, the multi-group cross section is performed. At this stage, a cross section is used as one of the infinite dilution, though in practical situations the foil is of certain thickness, and sometimes thick foil is adopted. When foil of certain thickness is put into the neutron field, the foil surface absorbs quickly the neutrons that are of large cross section of absorption. The shielding effect reduces the average effective cross- section of the foil. This is directly related to the accuracy of spectrum results, then, a shielding correction should be performed to the group cross-section for a foil of certain thickness.

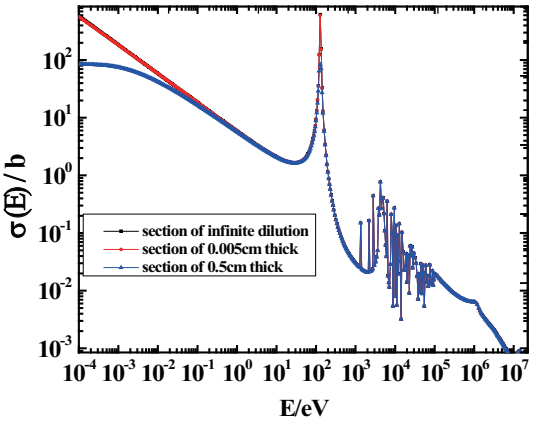

Fig. 1. (Color online) Shielding correction of cross section for ^{59}Co(n,γ)^{60}Co.

Considering a collimated neutron beam of one-way incidence, the cross section modification is to multiply each group of infinite dilute section with the shielding factor G_j for the group j [9].

$$G_j = \frac{1 - e^{-\tau_j}}{\tau_j} \qquad (6)$$

TABLE 1. Characteristic parameters of each foil and measured reaction rates

Nuclear reaction	Mass / mg	Abundance / %	Gamma energy / keV	γ_d / %	Reaction rate / s^{-1}
^{46}Ti(n,p)^{46}Sc	171.15	8	889.26	99.98	2.0465E-16
^{47}Ti(n,p)^{47}Sc	171.15	7.3	159.4	68	3.6430E-16
^{48}Ti(n,p)^{48}Sc	171.15	73.8	983.4	99.99	6.2666E-18
^{27}Al(n,α)^{24}Na	84.21	100	1368.55	100	1.4680E-17
^{63}Cu(n,γ)^{64}Cu	141.55	69.17	1345.76	0.49	7.2626E-15
^{59}Co(n,γ)^{60}Co	145.55	100	1332.51	99.98	4.7923E-14
^{54}Fe(n,p)^{54}Mn	264.05	5.9	834.81	99.98	1.3588E-15
^{56}Fe(n,p)^{56}Mn	264.05	91.72	846.6	99	2.2001E-17
^{24}Mg(n,p)^{24}Na	265.14	78.99	1368.55	100	2.9541E-17
^{58}Ni(n,p)^{58}Co	83.65	68.72	810.75	99.45	1.6775E-15
^{64}Zn(n,p)^{64}Cu	446.29	48.6	1345.76	0.49	6.1957E-16
^{115}In(n,γ)^{116m}In	0.09951	95.7	1293.54	85	2.2555E-13
^{45}Sc(n,γ)^{46}Sc	4.6560	100	1120.52	99.99	6.3933E-15
^{23}Na(n,γ)^{24}Na	2.551	100	1368.55	100	2.8021E-16
^{164}Dy(n,γ)^{165}Dy	8.805	28.2	361.67	0.84	2.6515E-13
^{176}Lu(n,γ)^{177}Lu	6.011	2.6	208.36	11	7.6517E-13
^{55}Mn(n,γ)^{56}Mn	0.2073	100	846.6	99	3.7069E-14
^{197}Au(n,γ)^{198}Au	0.06547	100	411.8	95.53	3.5251E-13
^{98}Mo(n,γ)^{99}Mo	22.65	24.13	140.46	90.9	1.3887E-14

where, $\tau_j = t\sum_{a,j}$ is the thickness of the foil making absorption length as the unit length, t is the thickness of the foil, and $\sum_{a,j}$ is the macroscopic absorption cross section of the group j. Taking an example of Co, effective group cross sections of three conditions are compared in Fig. 1.

B. Spectrum unfolding results of SAND-II and MSIT

The neutron energy range from 10^{-4} to 2×10^{7} eV was divided into 640 energy groups. The spectrum in the range 10^{-4}– 3.6×10^{-3} eV is set to 0, because most of neutrons in the range are shielded by the lead. The distribution of $\phi(E)\times E$ from 3.6×10^{-3} eV to 2×10^{7} eV was simulated using MCNP, shown in Fig. 2. The simulation result of $\phi(E)\times E - E$ was converted into $\phi(E) - E$ which was treated as the initial spectrum for SAND-II and MSIT. The initial spectrum and the results of spectrum unfolding were shown in Fig. 3. The calculated data of single action rates are given in Table 2, using the results of spectrum unfolding and Eq. (2).

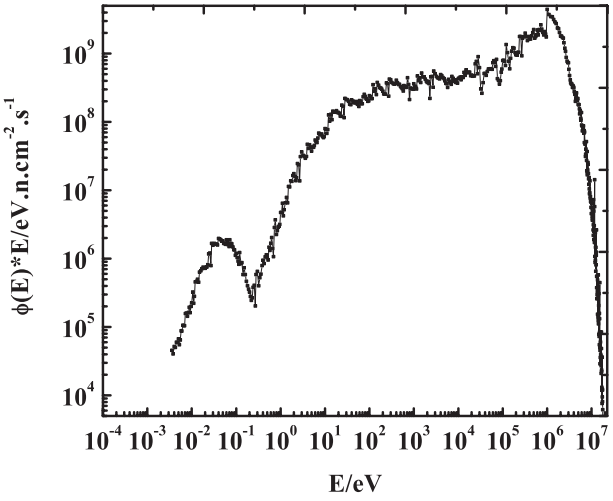

Fig. 2. Simulate ϕ (E) $\times$ E by MCNP.

V. DISCUSSION

Deviation of the neutron spectrum using the activation method includes the followings:

1) the measuring deviations of the single reaction rate. The deviation of any parameter in Eq. (5) affects the result of single reaction rate. The deviation of single reaction rate is about 3%–10%;

2) The deviation caused by group of cross-section;

3) The deviation caused by the initial spectrum;

4) The deviation caused by the process of spectrum unfolding. Selecting a set of good collocation foils helps the results come closer to the real neutron energy spectrum.

In Fig. 3, the results of spectrum unfolding using SAND-II and MSIT are nearly the same. From Table 2, the difference between the calculated and the measured data for ^{58}Ni(n,p)^{58}Co is big due to measurement error. The singular point of resonance region in Fig.3 is due to the measurement error of ^{58}Ni(n,p)^{58}Co.

The uncertainty analysis [10, 11] is made by MC in the following process [12]: 1) the error of single reaction rate and the uncertainty of cross section for each activation foil are given; 2), new reaction rate and nuclear cross section are formed using random sampling methods of extracting deviation with the original reaction rate and nuclear reaction cross section; and 3) the uncertainty of iterative solution spectrum is calculated.

TABLE 2. Results of calculated and measured reaction rates

Nuclear reaction [a]	A(I)	R_1(I)	δ_1(%)	R_2(I)	δ_2(%)
^{46}Ti(n,p)^{46}Sc	2.0465E-16	2.0135E-16	1.614	2.0146E-16	1.561
^{47}Ti(n,p)^{47}Sc	3.6430E-16	3.4574E-16	5.096	3.4479E-16	5.355
^{48}Ti(n,p)^{48}Sc	6.2666E-18	6.0531E-18	3.406	6.0539E-18	3.395
^{27}Al(n,α)^{24}Na	1.4680E-17	1.4817E-17	0.932	1.4839E-17	1.085
^{63}Cu(n,γ)^{64}Cu	7.2626E-15	7.2636E-15	0.014	7.2636E-15	0.014
^{59}Co(n,γ)^{60}Co	4.7923E-14	4.7923E-14	0.001	4.7923E-14	0.001
^{54}Fe(n,p)^{54}Mn	1.3588E-15	1.3368E-15	1.621	1.3347E-15	1.775
^{56}Fe(n,p)^{56}Mn	2.2001E-17	2.2409E-17	1.853	2.2373E-17	1.689
^{24}Mg(n,p)^{24}Na	2.9541E-17	2.9945E-17	1.369	2.9902E-17	1.221
^{58}Ni(n,p)^{58}Co	1.6775E-15	1.8554E-15	10.602	1.8510E-15	10.341
^{64}Zn(n,p)^{64}Cu	6.1957E-16	6.0565E-16	2.247	6.0535E-16	2.296
^{115}In(n,γ)^{116m}In	2.2555E-13	2.2557E-13	0.007	2.2557E-13	0.007
^{45}Sc(n,γ)^{46}Sc	6.3933E-15	6.3937E-15	0.006	6.3937E-15	0.006
^{23}Na(n,γ) ^{24}Na	2.8021E-16	2.8022E-16	0.004	2.8023E-16	0.005
^{164}Dy(n,γ)^{165}Dy	2.6515E-13	2.6514E-13	0.003	2.6514E-13	0.002
^{176}Lu(n,γ)^{177}Lu	7.6517E-13	7.6515E-13	0.002	7.6516E-13	0.001
^{55}Mn(n,γ)^{56}Mn	3.7069E-14	3.7069E-14	0.001	3.7069E-14	0.001
^{197}Au(n,γ)^{198}Au	3.5251E-13	3.5251E-13	0.001	3.5251E-13	0.0004
^{98}Mo(n,γ)^{99}Mo	1.3887E-14	1.3889E-14	0.016	1.3890E-14	0.018

[a] A(I) is the measured value of single reaction rate, R_1(I) is the calculated value of single reaction rate using the result of SAND-II; R_2(I) is the calculated value of single reaction rate using the result of MSIT, $\delta_1 = 100 \times |A(\mathrm{I}) - R_1(\mathrm{I})|/A(\mathrm{I})$, $\delta_2 = 100 \times |A(\mathrm{I}) - R_2(\mathrm{I})|/A(\mathrm{I})$

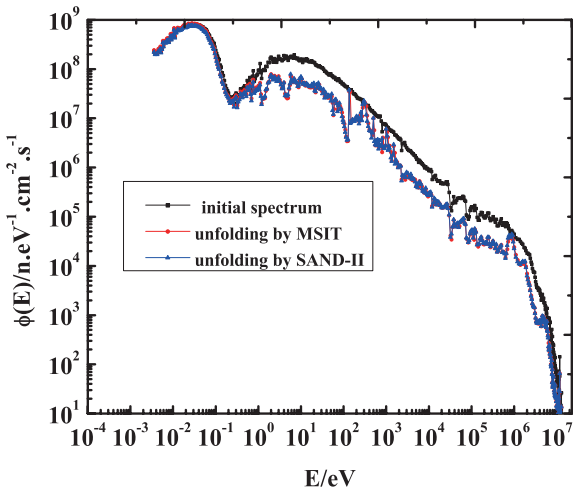

Fig. 3. (Color online) Unfolding results by SAND-II and MSIT.

Each group of relative deviation for the spectrum unfolding by SAND-II is shown in Fig. 4. The uncertainty of energy region from 10^{-4} eV to 10^{0} eV is $\pm 5\%$; the uncertainty of energy region from 10^{0} eV to 5×10^{4} eV is $\pm 30\%$; the uncertainty of energy region from 5×10^{4} eV to 2×10^{7} eV is $\pm 15\%$. From above analysis, the deviation of group cross section near the resonance peak is larger.

VI. CONCLUSION

Measurements made with foil-activation technique provided valuable information about the neutron spectrum in the irradiation chamber 3 of a research reactor. The group cross sections of a certain thickness of foil were processed before unfolding.

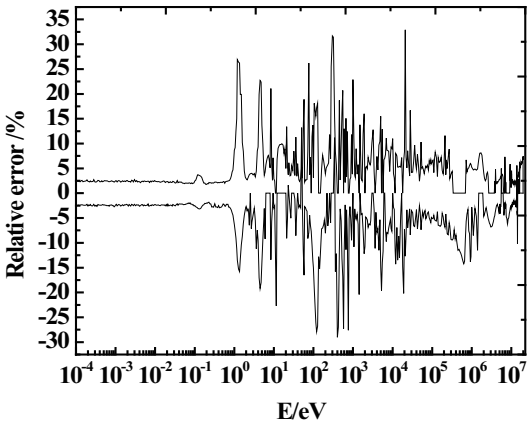

Fig. 4. Uncertainty of solution for SAND-II.

Two methods of spectrum unfolding based on iterative principle are used. The results of spectrum unfolding are generally consistent. The uncertainty analysis of result for spectrum unfolding was made by MC which was added to the process of SAND-II. If the result is not dependent on the initial spectrum, SAND-II will be perfect. From the above analysis, the experiment is successful due to the suitable foils and spectrum unfolding methods. One can know the neutron spectrum of irradiation chamber 3 that is important to the irradiation test in the future.

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